Andreev reflection off a fluctuating superconductor in the absence of equilibrium

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Andreev reflection between a normal metal and a superconductor whose order parameter exhibits quantum phase fluctuations is examined. The approach chosen is non perturbative in the tunneling Hamiltonian, and enables to probe the whole range of voltage biases up to the gap amplitude. Results are illustrated using the one–dimensional Josephson junction array model previously introduced in the linear response regime. Phase fluctuations are shown to affect the differential conductance and are compared to the result of Blonder, Tinkham and Klapwijk for a rigid BCS superconductor. The noise spectrum of the Andreev current is also obtained and its second derivative with respect to frequency is proposed as a direct tool to analyze the phase fluctuations.

I. INTRODUCTION

In the last decades, a considerable effort has been devoted towards the study of the transport properties of normal metal-superconductor (NS) junctions. The situation for the Andreev current and finite frequency noise is well understood when the superconductor is of the BCS type. More recent works have dealt with superconductors whose order parameter has a d-wave symmetry [1,2]. The role of collective modes arising from the phase of the fluctuations of the superconductor has also been addressed in the framework of linear response theory in high-T_c materials [3], as well as in one-dimensional array of Josephson junctions [4]. Some recent attempts have also included the effect of classical phase fluctuations of the order parameter on Andreev transport, either in the tunneling regime [5] or using Bogolubov-de Gennes equations [6].

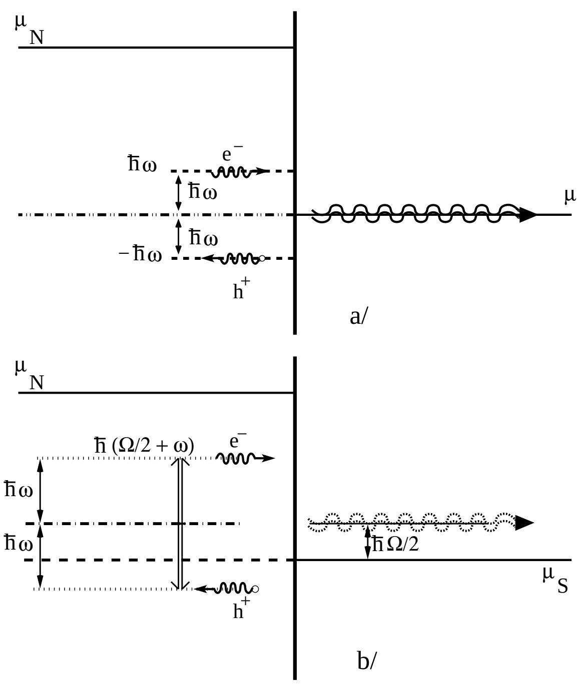

FIG. 1. Andreev reflection (bias voltage $eV = \mu_N - \mu_S$): a) In a rigid superconductor, the two electrons (one electron e^- and a time reversed hole h^+) have opposite energies $\pm\hbar\omega$ with respect to the superconductor chemical potential μ_S, and a Cooper pair is created on the right hand side. b) In the presence of phase fluctuations, the superconductor absorbs a collective mode with energy $\hbar\Omega$, so that the energies of the two incoming electrons are no more symmetrical with respect to μ_S.

A physical picture of the Andreev process is drawn in Fig. 1. A voltage bias is applied, such that the chemical potential of the metal μ_N lies eV above that of the superconductor μ_S. The origin of the energies is taken at μ_S. In Andreev reflection with conventional, rigid superconductors, Fig. 1a, an electron below μ_N, with an energy $\hbar\omega$ is reflected as a hole and a Cooper pair propagates in the superconductor. This pair has zero energy. The reflected hole can be interpreted as an electron below μ_S, with energy $-\hbar\omega$, which enters the superconductor. At zero temperature, the only possible values of $\hbar\omega$ are between $-eV$ and eV, see Fig. 1a , because both electrons states must have an energy below μ_N.

Turning to the case of a superconductor with phase fluctuations, Fig. 1b: exciting a collective mode in the superconductor costs an energy $\hbar\Omega$. One can therefore take an electron of energy $\hbar(\Omega/2+\omega)$ and reflect a hole, which is equivalent to a missing electron of energy$-\hbar(\omega-\Omega/2)$. The allowed values of $\hbar\omega$ must be smaller than $eV-\hbar\Omega/2$ (no electrons present above μ_N). Note that here, one has to sum over all possible values of $\hbar\Omega\in[0;2eV]$, taking care of the fact that this is a weighted sum, with a probability distribution $P(\Omega)$ which is determined by the Hamiltonian describing the phase.

In this paper, the Andreev current is calculated non-perturbatively in the tunneling Hamiltonian for N-S junctions whose Hamiltonian includes quantum phase fluctuations. The approach is inspired from the Keldysh technique of references [7,8], and enables the calculation of the current-voltage characteristics of the Andreev current when the bias voltage eV is close to (but smaller than) the gap amplitude $|\Delta|$. In particular, this corresponds to the experimental situation encountered in scanning tunneling microscope (STM) experiments performed on high-T_c materials [10–12]. Although the calculation is restricted here to an s-wave order parameter, qualitative features can be derived for transport in N-S junctions for arbitrary superconductors, provided that there is a gap. Here, a phase-only effective Hamiltonian will be used, which was previously derived in the literature [13]. In one dimension, this model is isomorphic to an array of Josephson junctions, with Josephson coupling energy E_J and charging energy E_0. The current and the finite frequency noise spectrum $S(\omega)$ are both derived, which requires to go beyond linear response theory. In particular, it will be shown that the second derivative of the noise with respect to the frequency $d^2S/d\omega^2$ gives a direct access to information about phase fluctuations, independently of the model chosen to describe the latter.

There are other situations where Andreev scattering is mediated by excitations at the normal supraconductor boundary. If the normal side is replaced by a ferromagnet [9], the spin of electrons pairs emerging from/entering the superconductor can suffer a spin flip accompanied by the emission/destruction of a magnon. This additional scattering channel effectively may enhance the Andreev current. Except for the case of a half-metal, electrons with opposite spins as well as electrons with the same spin then contribute to the Andreev current. The differences with our phase fluctuations calculation will addressed below.

The paper is organized as follows: in section II , the model Hamiltonian is discussed. In section III, the perturbative scheme which allows to derive the relevant Green's functions for the transport properties is described. In section IV, the phase fluctuations are introduced in this framework. The current-voltage characteristics associated with the linear Josephson junction array model of Ref. [4] for the phase appears in section V, as a function of the physical parameters E_J, E_0, as well as the barrier transparency.

II. MODEL HAMILTONIAN

The Hamiltonian describing the NS junction is composed of three terms: $H=H_N+H_t+H_{JJA}$. The first term is simply the Hamiltonian for the metal which is specified to a one band model $H_N=\sum_{k,\sigma}\epsilon_{k,\sigma}c^\dagger_{k,\sigma}c_{k,\sigma}$ with width W. The second term H_t is the tunneling term,

$$H_t=\sum_{N,S,\sigma}\Gamma c_{N,\sigma}c^\dagger_{S,\sigma}+h.c., \tag{1}$$

where Γ is a hopping amplitude which transfers electrons to/from the superconductor.

In the superconductor, it is assumed that the low energy lying excitations are given by fluctuations of the phase of the order parameter. If we forget the poorly screened long-range part of the Coulomb interaction, the Hamiltonian is the same as the one describing an array of Josephson junctions [4,14].

$$H_{JJA}=\sum_{i,j}E_J\cos(\theta_i-\theta_j)+\sum_i\frac{q_i^2}{2C}\ , \tag{2}$$

where E_J is the Josephson coupling (between islands i and j), the operator θ_i is the phase of the order parameter at "site" i, q_i is the charge on island i and $\frac{q_i^2}{2C}$ is the charging energy of island i. Eq. (2) can be put on a firmer

basis, starting from microscopic models. In particular, a phase-only Hamiltonian H_S has been derived from various microscopic models such as the attractive Hubbard model [13]:

$$H_S = \sum_{i,j} \frac{\hbar^2 N_S^0}{2m^*} \cos(\theta_i - \theta_j) + \sum_i \frac{1}{2} \frac{m^* v^2}{N_S^0} \rho_i^2 + \sum_{i,j} \frac{\pi e^2}{\varepsilon} \frac{\rho_i \rho_j}{|\, r_i - r_j \,|} \ , \tag{3}$$

where N_S^0 is the bare superfluid density, ε is the dielectric constant, $|\, r_i - r_j \,|$ is the distance between site i and j. The first term is the Josephson coupling between location i and location j. The second term is the local density fluctuation energy $(\rho - \langle\rho\rangle)^2/2\chi_0$, where $\langle\rho\rangle$ is the average density and χ_0 a density susceptibility. The last term is due to poorly screened long range Coulomb interactions. Since electron and hole-quasiparticles are not quasiparticles for the superconductor, the injected electrons and holes will decay and form pairs that will excite the eigenmodes of H_S.

III. NON–EQUILIBRIUM GREEN'S FUNCTIONS AND TRANSPORT

A voltage bias is introduced between the normal metal and the superconductor and the average current reads

$$I(t) = \frac{ie}{\hbar} \sum_\sigma \Gamma \langle c^\dagger_{N,\sigma}(t) c_{S,\sigma}(t) \rangle - h.c.. \tag{4}$$

Tunneling occurs from the bulk of the normal metal to the bulk of the superconductor through a narrow constriction, the tip of a STM for example.

The Keldysh formalism is used to express the current in terms of Green's functions. The definitions and properties of this system have been studied in detail in Ref. [8]. Here we adopt the same notations and start from their expression:

$$\langle I(t) \rangle = \frac{2e}{\hbar^2} \,|\, \Gamma \,|^2 \int_{-\infty}^{\infty} \left[g^{+-}_{NN11}(t,t_1) G^{-+}_{SS11}(t_1,t) - g^{-+}_{NN11}(t,t_1) G^{+-}_{SS11}(t_1,t) \right] dt_1. \tag{5}$$

The dressed Keldysh Green matrices on the superconducting side are expanded using the Dyson expansion

$$\hat{G}^{\pm\mp}_{SS}(t,t') = \left[\hat{G}^r_{SS} \hat{\Sigma}^\dagger \hat{g}^{\pm\mp}_{NN} \hat{\Sigma} \hat{G}^a_{SS} \right](t,t'), \tag{6}$$

where $\hat{\Sigma}$ is the self energy matrix describing the hopping from the normal side to the superconductor, and $\hat{\Sigma}^\dagger$ describes the opposite process. For simplicity, integrals over time have been omitted. Note that only subgap tunneling is considered here, allowing to set $\hat{g}^{\pm\mp}_{SS} = 0$. Inserting the Dyson expansion, the current reads:

$$\begin{aligned} \langle I(t) \rangle = \frac{2e}{\hbar^4} |\Gamma|^4 & \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} G^r_{SS12}(t_1,t_2) G^a_{SS21}(t_3,t) \times \\ & \times \Big(g^{+-}_{NN11}(t,t_1) g^{-+}_{NN22}(t_2,t_3) - g^{-+}_{NN11}(t,t_1) g^{+-}_{NN22}(t_2,t_3) \Big) dt_1 dt_2 dt_3 \ . \end{aligned} \tag{7}$$

The dressed, retarded Green's function on the superconducting side is given by:

$$G^r_{SS12}(t',t) = g^r_{SS12}(t',t) + \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \Big(\hat{g}^r_{SS}(t',t_1) \hat{\Sigma}^r_S(t_1,t_2) \hat{G}^r_{SS}(t_2,t) \Big)_{12} dt_1 dt_2 + \ldots, \tag{8}$$

with the dressed self energy $\hat{\Sigma}^r_S$ defined as:

$$\hat{\Sigma}^r_S(t_1,t_2) = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \hat{\Sigma}^*(t_1,t_1') \hat{g}^r_{NN}(t_1',t_2') \hat{\Sigma}(t_2',t_2) dt_1' dt_2' \ , \tag{9}$$

and the unperturbed momentum averaged Green's function in the superconductor reads :

$$g^r_{SS12}(\omega) = \frac{\hbar}{W} \frac{|\, \Delta \,|}{\sqrt{|\, \Delta \,|^2 - (\hbar\omega + i\alpha)^2}} e^{i\theta_0} \equiv \tilde{g}^r_{SS12}(\omega) e^{i\theta_0}, \tag{10}$$

where α is an infinitesimally small positive quantity and θ_0 the phase of the superconductor, which appears also in the gap parameter: $\Delta = |\, \Delta \,|\, e^{i\theta_0}$.

IV. TRANSPORT IN THE PRESENCE OF PHASE FLUCTUATIONS

In the presence of phase fluctuations, the Green's function on the superconducting side contains two time scales [15]. First, fast time scales (of the order $\hbar/\Delta$) which were described in the previous section in Eq. (10). Secondly, "slow" time scales associated with the fluctuating phase $\theta(x,t)$:

$$\Delta = \mid \Delta \mid e^{i\theta(x,t)}. \tag{11}$$

The phase of the order parameter has both spatial and temporal variations and is no longer equal to $\theta_0 = \theta(0,0)$. Tunneling is supposed to occur close to the interface. The typical dimension parallel to the interface will be small with respect to both the classical correlation length ξ_ϕ^c [16] and to the amplitude correlation length ξ_a. In high-T_c superconductors, ξ_ϕ^c and ξ_a are of the order of a few nanometers. ξ_ϕ^c is believed to be generally not much larger than ξ_a, and ξ_a is much smaller than in BCS superconductors. We shall thus replace x by zero. The temporal variations of θ are due to quantum fluctuations; the phase of the order parameter and the number of pairs are conjugated variables. These variations occur on time scales of the order at least of the plasma frequency $\omega_p \equiv \sqrt{8E_0E_J}/\hbar$ with $E_0 = q_0^2/2C$ and E_J appear in the definition of the Hamiltonian of Eq. (2). Equivalently, $\hbar\omega_p <\mid \Delta \mid$, which is the contrary to what usually occurs in BCS superconductors. The dynamic nature of the phase is reflected in the anomalous Green's functions [5]:

$$g^{r,a}_{SS12}(t',t) = \tilde{g}^{r,a}_{SS12}(t',t)e^{i\frac{1}{2}\left(\theta(0,t)+\theta(0,t')\right)} \tag{12}$$

where $\tilde{g}^{r,a}_{SS12}(t',t)$ is the inverse Fourier transform of $\tilde{g}^{r,a}_{SS12}(\omega)$, and $g^{r,a}_{SS21}(t',t)$ has the opposite phase. Because of the slow temporal variation of the phase, the exponential in Eq. (12): $\exp[i(\theta(0,t)+\theta(0,t'))/2] \approx \exp[i\theta(0,t')]$ without affecting the results in a significant manner.

A. Current and differential conductance

Substituting Eq. (12) in the current, the fourth (lowest non vanishing) order term in Γ which contributes to the current is:

$$\begin{aligned}\langle I(t)\rangle = \frac{2e}{\hbar^4}\mid\Gamma\mid^4 \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \tilde{g}^a_{SS21}(t_3,t)\tilde{g}^r_{SS12}(t_1,t_2)\langle e^{i\theta(0,t_1)}e^{-i\theta(0,t_3)}\rangle \\ \times\Big(g^{+-}_{NN11}(t,t_1)g^{-+}_{NN22}(t_2,t_3) - g^{-+}_{NN11}(t,t_1)g^{+-}_{NN22}(t_2,t_3)\Big)dt_1dt_2dt_3.\end{aligned} \tag{13}$$

The bracket $\langle ...\rangle$ signifies that an average has been performed over the dynamical degrees of freedom of the phase. Depending on the nature of the phase, this will correspond to a Gaussian model – the so called ordered phase – or to a nonlinear model [17] (see below). Note that this expression is analogous to the one derived several decades ago by Combescot *et al.* for inelastic tunneling [18]: there the working assumption was that the inelastic coupling did not couple directly the two electrodes. Here, this same assumption is natural because the phase fluctuations happen only on the superconducting side. The correlator of the phase and its Fourier transform are denoted:

$$p(t) = \langle e^{i\theta(0,t)}e^{-i\theta(0,0)}\rangle \ , \tag{14a}$$

$$P(\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{i\omega t}p(t)dt \ . \tag{14b}$$

$P(\omega)$ can also be viewed as the occupation probability of the collective phase modes with energy $\hbar\omega$. Using the Fourier representation, the current becomes:

$$\begin{aligned}\langle I(t)\rangle = \frac{2e}{h\hbar^3}\mid\Gamma\mid^4 \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \tilde{g}^a_{SS21}(\omega+\Omega/2)\tilde{g}^r_{SS12}(\omega-\Omega/2) \\ \times\Big[g^{+-}_{NN11}(\omega+\Omega/2)g^{-+}_{NN22}(\omega-\Omega/2) - g^{-+}_{NN11}(\omega+\Omega/2)g^{+-}_{NN22}(\omega-\Omega/2)\Big]P(\Omega)d\Omega d\omega.\end{aligned} \tag{15}$$

Next, for frequencies below the gap, one can approximate $\tilde{g}^{a,r}_{SSij}(\omega\pm\Omega/2) \simeq \tilde{g}^{a,r}_{SSij}(\omega) \simeq \hbar/\left(W\sqrt{1-\hbar^2\omega^2/|\Delta|^2}\right)$ (W is the bandwidth), and the current becomes:

$$
\begin{aligned}
\langle I(t)\rangle &= \frac{2e}{h\hbar^3}\,|\Gamma|^4\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\tilde{g}^a_{SS21}(\omega)\tilde{g}^r_{SS12}(\omega)\\
&\times\Big[g^{+-}_{NN11}(\omega+\Omega/2)g^{-+}_{NN22}(\omega-\Omega/2)-g^{-+}_{NN11}(\omega+\Omega/2)g^{+-}_{NN22}(\omega-\Omega/2)\Big]P(\Omega)d\Omega d\omega,
\end{aligned}
\tag{16}
$$

where the two causal Green function elements in Eq. (16) are related to the electron density of states $\rho(\hbar\omega)$:

$$
g^{+-}_{NN11}(\omega) = 2\pi i\hat{\rho}_{11}(\hbar\omega-eV)n_F(\hbar\omega-eV)\ , \tag{17a}
$$

$$
g^{-+}_{NN22}(\omega) = 2\pi i\hat{\rho}_{22}(\hbar\omega+eV)\big[n_F(\hbar\omega+eV)-1\big]\ , \tag{17b}
$$

with $\hat{\rho}_{11}(\hbar\omega-eV)\equiv\hat{\rho}_{22}(-\hbar\omega+eV)\equiv\rho(\hbar\omega)\simeq\hbar/\pi W$.

Using the property $\tilde{g}^a_{SS21}(\omega)=[\tilde{g}^r_{SS12}(\omega)]^*$, the current in the absence of phase fluctuation is recovered by setting $P(\Omega)$ to be a delta function:

$$
\langle I\rangle_0 = \frac{8e}{h\hbar}\frac{|\Gamma|^4}{W^2}\int_{-\infty}^{\infty}|\,\tilde{g}^a_{SS21}(\omega)\,|^2\,\Big[n_F(\hbar\omega-eV)-n_F(\hbar\omega+eV)\Big]d\omega, \tag{18}
$$

where subscript 0 means that in this limit $E_0/E_J=0$. The known result for the Andreev current is recovered [8]. It is well known however that even in the absence of phase fluctuations, a perturbative calculation to fourth order in Γ is not sufficient to give a satisfactory answer for the whole range of voltage biases between 0 and Δ. Indeed, a resummation procedure, explicited by Eq. (8), has to be carried out. In the absence of phase fluctuations, it is thus sufficient to replace $\tilde{g}^r_{SS21}(\omega)$ by $G^r_{SS21}(\omega)$: this allows to recover the scattering theory results of BTK [19].

Turning back to the lowest order contribution of the current in the presence of phase fluctuations, one obtains:

$$
\begin{aligned}
\langle I\rangle &= \frac{8e}{h\hbar}\frac{|\Gamma|^4}{W^2}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}G^a_{SS21}(\omega)G^r_{SS12}(\omega)\\
&\times\Big[n_F(\hbar\omega-eV+\hbar\Omega/2)-n_F(\hbar\omega+eV-\hbar\Omega/2)\Big]P(\Omega)d\Omega d\omega.
\end{aligned}
\tag{19}
$$

Note that $P(\Omega)$ decreases when Ω becomes large. In particular, the integrated contribution $\int_\Omega^\infty P(\Omega')d\Omega'$ becomes negligible well *before* $\hbar\Omega>2eV$, allowing modify the upper bound of the integral over Ω. Simultaneously, the advanced (retarded) Green's functions are dressed as before and take the form :

$$
G^{r,a}_{SS12}(\omega) = \frac{1}{D^{r,a}(\omega)}\tilde{g}^{r,a}_{SS12}(\omega), \tag{20}
$$

so that at zero temperature the current reads:

$$
\langle I\rangle \simeq \frac{16e|\Gamma|^4}{h\hbar W^4}\,|\,\Delta\,|^2\int_0^{2\frac{eV}{\hbar}}P(\Omega)d\Omega\int_0^{\frac{eV}{\hbar}-\frac{\Omega}{2}}\frac{1}{D(\omega)}d\omega, \tag{21}
$$

with

$$
\begin{aligned}
D(\omega) &= \left(\frac{|\,\Delta\,|^2}{\hbar^2}-\omega^2\right)D^r(\omega)D^a(\omega) = \left(\frac{|\,\Delta\,|^2}{\hbar^2}-\omega^2\right)|D^a(\omega)|^2\\
&= \left(\frac{|\,\Delta\,|^2}{\hbar^2}-\omega^2\right)\left(1+\frac{|\Gamma|^4}{W^4}\right)^2+4\frac{|\Gamma|^4}{W^4}\omega^2.
\end{aligned}
\tag{22}
$$

In a more rigorous approach, the expansions of $G^r_{SS12}(\omega)$ have to be performed in the presence of phase fluctuations. This means that they should contain not only linear terms in $P(\Omega)$ but also higher order correlators of the exponentiated phases. Here, in the expression for the current in Eq. (21), only the first order in $P(\Omega)$ has been retained, which constitutes the analog of a weak inelastic coupling – single phonon – approximation of Ref. [18].

The differential conductance $d\langle I\rangle/dV$ can be straightforwardly computed from Eq. (21):

$$
\frac{d\langle I\rangle}{dV} \simeq \frac{16e^2|\Gamma|^4}{h\hbar^2 W^4}\,|\,\Delta\,|^2\int_0^{2\frac{eV}{\hbar}}P(y)\frac{1}{D\left(\frac{eV}{\hbar}-\frac{y}{2}\right)}dy. \tag{23}
$$

B. Finite frequency noise

Another useful tool which probes the fluctuations is the finite frequency noise. The symmetrized real time noise correlator is given by:

$$S(t-t') = \langle \delta I(t)\, \delta I(t') \rangle + \langle \delta I(t')\, \delta I(t) \rangle \tag{24}$$

where $\delta I(t) = I(t) - \langle I(t) \rangle$. This current–current correlator is expressed in terms of the Keldysh Green's functions: using the self energy matrix, we can write it as a trace.

$$\begin{aligned} S(t,0) = -e^2 \mathrm{Tr}\Big[\Big\{ \Big(& \hat{\sigma}_z \hat{\Sigma} \hat{G}^{+-}_{SN}(0,t) \hat{\sigma}_z \hat{\Sigma} \hat{G}^{-+}_{SN}(t,0) - \hat{\sigma}_z \hat{\Sigma}^\dagger \hat{G}^{+-}_{NN}(0,t) \hat{\sigma}_z \hat{\Sigma} \hat{G}^{-+}_{SS}(t,0) \Big) + \Big(\ \hat{\Sigma} \leftrightarrow \hat{\Sigma}^\dagger\ ;\ S \leftrightarrow N\ \Big) \Big\} \\ & + \{\ (0,t) \leftrightarrow (t,0)\ \} \Big], \end{aligned} \tag{25}$$

where $\hat{\sigma}_z$ is a Pauli matrix.

The lowest non vanishing contribution (order 4 in Γ) is extracted by expanding the Green's functions in Eq. (25). The corresponding Dyson equations read (time integrations are implicit):

$$\begin{aligned} \hat{G}^{\pm\mp}_{SN} &= \hat{G}^r_{SS} \hat{\Sigma}^\dagger \hat{g}^{\pm\mp}_{NN}\ , \\ \hat{G}^{\pm\mp}_{NS} &= \hat{g}^{\pm\mp}_{NN} \hat{\Sigma} \hat{G}^a_{SS}\ , \\ \hat{G}^{\pm\mp}_{NN} &= \hat{g}^{\pm\mp}_{NN}\ , \end{aligned} \tag{26}$$

where we have purposely written the Green's functions to the lowest order which they contribute. Inserting these expressions in the real time correlator gives:

$$\begin{aligned} S(t,0) = -e^2 \int_{-\infty}^{+\infty} dt_1 \int_{-\infty}^{+\infty} dt_2 \mathrm{Tr}\Big[\Big\{ & \hat{\sigma}_z \hat{\Sigma} \hat{G}^r_{SS}(0,t_1) \hat{\Sigma}^\dagger \hat{g}^{+-}_{NN}(t_1,t) \hat{\sigma}_z \hat{\Sigma} \hat{G}^r_{SS}(t,t_2) \hat{\Sigma}^\dagger \hat{g}^{-+}_{NN}(t_2,0) \\ & + \hat{\sigma}_z \hat{\Sigma}^\dagger \hat{g}^{+-}_{NN}(0,t_1) \hat{\Sigma} \hat{G}^a_{SS}(t_1,t) \hat{\sigma}_z \hat{\Sigma}^\dagger \hat{g}^{-+}_{NN}(t,t_2) \hat{\Sigma} \hat{G}^a_{SS}(t_2,0) \\ & - \hat{\sigma}_z \hat{\Sigma} \hat{G}^r_{SS}(0,t_1) \hat{\Sigma}^\dagger \hat{g}^{+-}_{NN}(t_1,t_2) \hat{\Sigma} \hat{G}^a_{SS}(t_2,t) \hat{\sigma}_z \hat{\Sigma}^\dagger \hat{g}^{-+}_{NN}(t,0) \\ & - \hat{\sigma}_z \hat{\Sigma}^\dagger \hat{g}^{+-}_{NN}(0,t) \hat{\sigma}_z \hat{\Sigma} \hat{G}^r_{SS}(t,t_1) \hat{\Sigma}^\dagger \hat{g}^{-+}_{NN}(t_1,t_2) \hat{\Sigma} \hat{G}^a_{SS}(t_2,0) \Big\} + \Big\{ 0 \leftrightarrow t \Big\} \Big]. \end{aligned} \tag{27}$$

The procedure for including the phase fluctuations in the noise correlator is identical to that used in the expression of the current of Eq. (7). However, here additional contributions which are proportional to $\hat{G}^r_{SS} \hat{G}^r_{SS}$ or to $\hat{G}^a_{SS} \hat{G}^a_{SS}$ occur, but these turn out not contribute to the noise to lowest order in the phase correlator.

The current noise spectrum $S(\omega)$ is the Fourier transform of $S(t)$. In the subgap regime, the only non–vanishing elements of the causal Green's functions are the off diagonal ones : $\tilde{g}^{a,r}_{SS12} = \tilde{g}^{a,r}_{SS21} \simeq \hbar/W$. The current noise spectrum can be expressed in terms of the off diagonal Keldysh Green's functions:

$$\begin{aligned} S(\omega) \simeq \frac{4e^2}{\hbar^2} \frac{|\Gamma|^4}{2\pi W^2} \int_0^{2\frac{eV}{\hbar}} & d\Omega P(\Omega) \int_{-\infty}^{+\infty} d\omega_1 \\ & \Big[g^{+-}_{N22}(\omega_1 - \Omega) g^{-+}_{N11}(\omega_1 - \omega) \\ & + g^{+-}_{N11}(\omega_1 + \Omega) g^{-+}_{N22}(\omega_1 - \omega) \\ & + g^{+-}_{N11}(\omega_1 + \Omega) g^{-+}_{N22}(\omega_1 + \omega) \\ & + g^{+-}_{N22}(\omega_1 - \Omega) g^{-+}_{N11}(\omega_1 + \omega) \Big]. \end{aligned} \tag{28}$$

The integration over ω_1 defines the relevant energy intervals which contribute to the noise:

$$S(\omega) \simeq \frac{16e^2}{2\pi} \frac{|\Gamma|^4}{W^4} \Big[\int_0^{2\frac{eV}{\hbar} - \omega} d\Omega P(\Omega) \Theta\left(\frac{2eV}{\hbar} - \omega - \Omega \right) \left(\frac{2eV}{\hbar} - \omega - \Omega \right) + \int_0^{2\frac{eV}{\hbar} + \omega} d\Omega P(\Omega) \left(\frac{2eV}{\hbar} + \omega - \Omega \right) \Big], \tag{29}$$

where $\Theta(\Omega)$ is the Heaviside function.

In the limit of low frequencies, the noise becomes:

$$S(\omega=0) = \frac{32e^2}{2\pi}\frac{|\Gamma|^4}{W^4}\int_0^{2\frac{eV}{\hbar}} d\Omega P(\Omega)\left(\frac{2eV}{\hbar}-\Omega\right)$$
$$= 4e\langle I\rangle, \tag{30}$$

which corresponds to the Schottky formula. Note that this constitutes an illustration of Schottky formula for a situation where the charge transfer is effectively inelastic: as shown in Fig. 1b, the transfer of two electrons in the superconductor generates phase quanta and thus constitutes an inelastic process. These processes are included when the distribution $P(\omega)$ in the current and noise deviates from a delta function.

The current noise spectrum can be measured experimentally and provides a direct way to obtain information on $P(\omega)$ and thus on phase fluctuations. In the tunnel limit, the distribution $P(\omega)$ is related to the second derivative of the current noise spectrum by the formula

$$\frac{d^2S}{d\omega^2} \simeq \frac{\hbar\langle I\rangle_0}{V}\left[P\left(\frac{2eV}{\hbar}+\omega\right)+P\left(\frac{2eV}{\hbar}-\omega\right)\Theta\left(\frac{2eV}{\hbar}-\omega\right)\right]. \tag{31}$$

The prefactor stands for the current in the absence of phase fluctuations :$\langle I\rangle_0 = (16e^2V|\Gamma|^4)/(\hbar W^4)$.

Note that so far, no specific model for the phase fluctuations has been specified, as the only assumption used what the fact that $P(\Omega)$ falls sufficiently rapidly under the gap. In the next section, current voltage characteristics are plotted using a specific model to describe the phase dynamics.

V. APPLICATION TO THE 1D JOSEPHSON-JUNCTION ARRAY MODEL

One spatial dimension is specified, motivated by the fact that tunneling typically occurs near the tip of a scanning tunneling microscope. The dynamics of the phase can then be modeled by a one-dimensional Josephson-junction Array (JJA), which was previously studied in linear response [4]. The phase correlator in Eq. (14a) is then known. The parameters of this model are the Josephson energy E_J and the charging energy E_0 (which in turn define the plasma frequency), together with the parameter κ which depends on the ratio of the junction capacitance divided by the capacitance of the islands. If the latter capacitance dominates over that of the junction, $\kappa = 2$.

The importance of phase fluctuations is monitored by the ratio $\hbar\omega_p/E_J$. In particular, for $\hbar\omega_p/E_J < \pi/2$, the phase Hamiltonian can be mapped to a Luttinger liquid model and $P(\Omega)$ can be derived in a standard way from harmonic oscillator correlators. On the other hand, for $\hbar\omega_p/E_J > \pi/2$, the phase correlator decays exponentially in time. This is identified as the disordered phase. At $\hbar\omega_p/E_J = \pi/2$, there is a Kosterlitz-Thouless transition between the ordered and the disordered phase.

A. Ordered phase

At zero temperature, the time dependent phase correlator is given by:

$$p(t) = \exp\left\{-\frac{\sqrt{2}}{\pi}\sqrt{\frac{E_0}{E_J}}\Big[\int_0^{\kappa\omega_p|t|}\frac{1-\cos x}{x}\frac{dx}{\sqrt{1-(x/\kappa\omega_p t)^2}} + i\,\mathrm{sgn}(t)\int_0^{\kappa\omega_p|t|}\frac{\sin x}{x}\frac{dx}{\sqrt{1-(x/\kappa\omega_p t)^2}}\Big]\right\}, \tag{32}$$

with sgn(t) the sign function. Its Fourier transform $P(\Omega)$ is identically zero for $\Omega < 0$. For $\Omega > 0$, it has a power law behavior close to $\Omega = 0$:

$$P(\Omega) \simeq \Omega^{-\left(1-\frac{1}{\pi}\sqrt{\frac{2E_0}{E_J}}\right)}, \tag{33}$$

and goes rapidly to zero for $\Omega > \kappa\omega_p$.

For $\Omega = \kappa\omega_p$, $P(\Omega)$ shows a weaker singularity [4] and diverges as $(\omega_p - \Omega)^{-\alpha}$, with $\alpha = 1/2 - \pi^{-1}\sqrt{2E_0/E_J}$ (here this corresponds to fixing the parameter of Ref. [4] $\kappa = 2$).

Turning to the transport properties, the effect of the phase fluctuations on the noise are probed. For convenience, we compare our results to those derived with a BTK model [19], which corresponds to an interface with a rigid superconductor. The quantity $\gamma = \Gamma/W$ corresponds to the transparency of the barrier. γ enters the prefactor of the delta function potential of BTK theory with the dependence $\hbar^2k_F(1-\gamma^2)/2m\gamma$ (k_F is the Fermi wave vector). $\gamma = 1$ corresponds to a perfect contact, while $\gamma \to 0$ describes an opaque barrier.

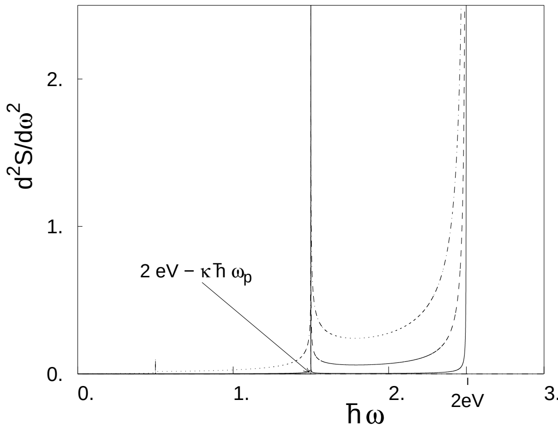

FIG. 2. Second derivative of the symmetrized noise spectrum versus frequency (in units of $4(e^2/h)\hbar\gamma^4/\omega_p$) for different values of $\hbar\omega_p/E_J$, $\hbar\omega_p/E_J = 0.01$ (solid line), $\hbar\omega_p/E_J = 0.2$ (dashed line), $\hbar\omega_p/E_J = 1$ (dashed dotted line). The plasma frequency ω_p is set to $|\Delta|/2\hbar$.

In Fig. 2 the second derivative of the symmetrized noise spectrum $d^2S/d\omega^2$ is plotted as a function of ω, for a perfectly good contact. Several values of the parameter $\hbar\omega_p/E_J$ are considered, starting from weak fluctuations to substantial collective excitations within the ordered phase. To illustrate the effect of phase fluctuations, the choice $\hbar\omega_p = |\Delta|/2$ is made, so that all the features in $P(\Omega)$ are located within the superconducting gap.

In the absence of phase fluctuations, the finite frequency noise has a singular derivative at $\hbar\omega = 2eV$ [20]. In Fig. 2, this would imply a delta function peak in $d^2S/d\omega^2$. For low voltages, there is no deviation with respect to the results of scattering theory. The effect of phase fluctuations is twofold: first, the delta function broadens to a power law divergence (partially cut on the figure), and second, it acquires a secondary peak – previously discussed for $P(\Omega)$ – at $2eV - \kappa\hbar\omega_p$. This illustrates the result of Eq. (31): an experimental measurement of the current noise spectrum constitutes a direct diagnosis of the importance of phase fluctuations.

Naturally, the differential conductance $d\langle I\rangle/dV$ in Eq. (23) is also affected by the phase fluctuations. It is studied below for different regimes: first, it is plotted as a function of bias, for both the case of a perfect contact and for the case of a weak transmission; second, the transparency is varied while the bias is fixed to a large value in order to explore the deviations from the tunnel limit. In each case, curves are obtained for several values of the ratio $\hbar\omega_p/E_J$ which characterizes the importance of the fluctuations.

The differential conductance of a perfect junction is displayed in Fig. 3 as a horizontal line at $d\langle I\rangle/dV = 4e^2/h$ [19]. In the presence of fluctuations, it saturates to the BTK value for biases larger than $\hbar\omega_p/2$ (here the bias is varied from 0 to $|\Delta|$): for sufficiently large eV, $\int_0^{2eV/\hbar} P(\Omega)d\Omega$ is essentially equal to unity because for $\gamma = 1$, $D(x)$ which enters Eq. (23) is a constant. Phase fluctuations have a tendency to decrease the differential conductance at low bias. In particular for $\hbar\omega_p/E_J \geq 1$, the significant contributions from the integral $\int_{\omega_p}^{2eV/\hbar} P(\Omega)d\Omega$ are the cause for the deviations from the ideal conductance at large bias.

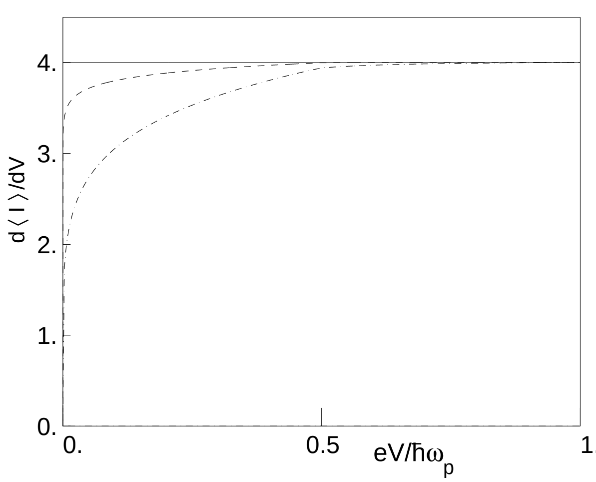

FIG. 3. Differential conductance $d\langle I\rangle/dV$ in units of e^2/h versus the ratio $eV/\hbar\omega_p$. A perfect contact ($\gamma = 1$) is considered and different values of $\hbar\omega_p/E_J$ are displayed : $\hbar\omega_p/E_J = 0$ (BTK curve, solid line), $\hbar\omega_p/E_J = 0.2$ (dashed line) and $\hbar\omega_p/E_J = 1$ (dash-dotted line).

The case of $\gamma = 1$ is somewhat academic because perfect contacts are difficult to achieve in practice. The same curves are plotted now for $\gamma = 0.4$ in Fig. 4. For intermediate biases, phase fluctuations do not affect the differential conductance drastically: at $eV = \hbar\omega_p$ for instance, $d\langle I\rangle/dV$ is essentially constant (of the order $4e^2\gamma^2/h$), and all curves can not be distinguished. However, deviations occur both at small and large voltages. For small voltages, according to Eq. (23) and due to the fact that $P(0) = 0$ the differential conductance in the presence of *any* phase fluctuation is required to vanish (this is not really obvious in Fig. 4 because of the choice of scale). More dramatic is the fact that phase fluctuations play a role at large voltages, contrary to the high transparency regime.

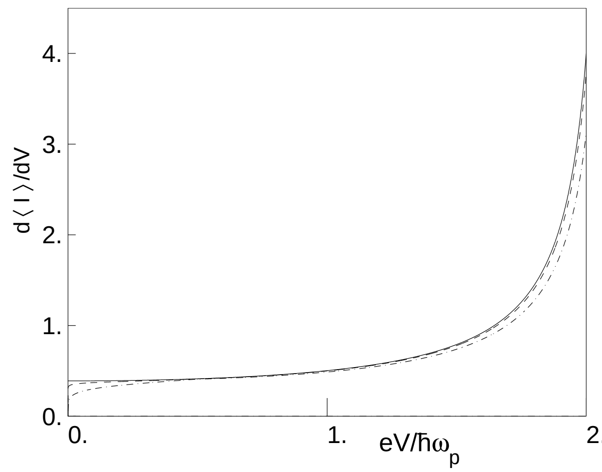

FIG. 4. Same as Fig. 3, for a transparency $\gamma = 0.4$.

In Fig. 5, the transparent regime is examined with a bias voltage $eV = |\Delta|$. In contrast to the previous curves, the differential conductance is plotted versus the transparency of the barrier. The plasma frequency and the ratio $\hbar\omega_p/E_J$ are chosen as previously.

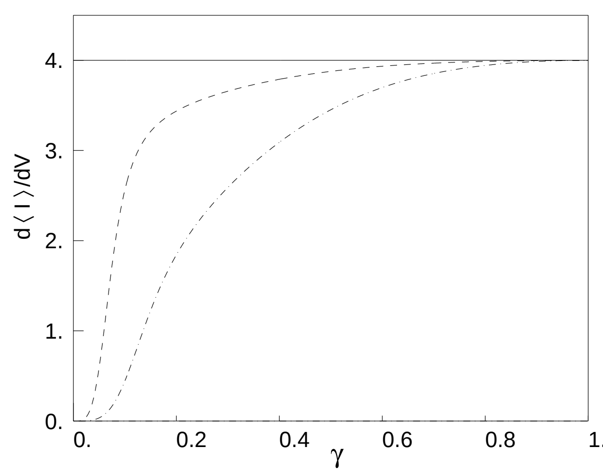

FIG. 5. Differential conductance (in units of e^2/h) as a function of the transparency, for a voltage bias $eV = |\Delta|$ (same convention for $\hbar\omega_p/E_J$ as in Fig. 3)

For barrier transparencies smaller than 0.1, the differential conductance has power law dependence ($d\langle I\rangle/dV \sim \gamma^4$). As the transparency is increased, in the presence of phase fluctuations an inflexion point appears and the rise of $d\langle I\rangle/dV$ is then slower. Note once again that it is reduced from the BTK case when phase fluctuations become important. As indicated previously in Fig. 3, the differential conductance saturates to the BTK value at high γ regardless of the degree of phase fluctuations.

B. Disordered phase

In the disordered phase ($\hbar\omega_p/E_J > \pi/2$), the behavior of the Fourier transform of the time dependent phase correlator close to the transition has been previously derived from the XY model [17]. For small Ω,

$$P(\Omega) \sim \Theta(\Omega - \omega_p\xi^{-1})(\Omega - \omega_p\xi^{-1})^{-1/2}, \tag{34}$$

where $\Theta(x)$ is the Heaviside function. The results of linear response theory of Ref. [4] are briefly recalled and compared to the predictions of the Keldysh calculation of Eq. (21). In this phase, a threshold voltage $\hbar\omega_p\xi^{-1}$ has to be reached in order to obtain a non zero current. Here, the Kosterlitz-Thouless correlation length (in dimensionless units) reads $\xi = \exp\left[b\left(2/\pi - \sqrt{E_J/8E_0}\right)^{-1/2}\right]$, where b is a positive constant. Because the goal is to predict the current as a function of voltage for both the tunnel limit and the transparent regime (for $0 < eV <\mid\Delta\mid$), $P(\Omega)$ needs to be characterized. $P(\Omega)$ can be set to zero for Ω larger than $\kappa\omega_p$. This assumption is reasonable below the Kosterlitz-Thouless transition. In accordance with Eq. (34), we adopt the form

$$P(\Omega) \simeq \frac{1}{2}((\kappa - \xi^{-1})\omega_p)^{-1/2}\Theta(\Omega - \omega_p\xi^{-1})\Theta(\kappa\omega_p - \Omega)(\Omega - \omega_p\xi^{-1})^{-1/2}, \tag{35}$$

Using Eq. (21) and (35), calculations can be performed analytically.

For $eV < \hbar\omega_p\xi^{-1}/2$, the average current vanishes. For $\xi^{-1} < 2eV/\hbar\omega_p < \kappa$, the average current reads :

$$\langle I\rangle = \frac{e}{h\hbar}\frac{16\gamma^4}{(1+\gamma^4)^2}\frac{\kappa\omega_p}{2}(1-(\kappa\xi)^{-1})\left(\frac{2\frac{eV}{\hbar\omega_p} - \xi^{-1}}{\kappa - \xi^{-1}}\right)^{3/2} f(\mathcal{A}), \tag{36}$$

where

$$f(x) = \frac{2}{x}\Big\{\frac{1}{2}\sqrt{1+\frac{2}{x}}\ \ln\Big|\frac{1+\sqrt{1+\frac{2}{x}}}{1-\sqrt{1+\frac{2}{x}}}\Big| - \sqrt{\frac{2}{x}-1}\ \arctan\left(\frac{1}{\sqrt{\frac{2}{x}-1}}\right)\Big\}\ , \tag{37a}$$

$$\mathcal{A} = \left(\frac{2eV}{\hbar\omega_p} - \xi^{-1}\right)\left(\frac{\hbar\omega_p}{|\Delta|}\right)\sqrt{1-\frac{4\gamma^4}{(1+\gamma^4)^2}}\ , \tag{37b}$$

where $\mathcal{A}$ is a scaling variable which varies between 0 (perfect contact $\gamma = 1$) and 1 ($\gamma = 0$ and a plasma frequency $\omega_p = |\Delta|/\hbar$). Note that the function f does not vary substantially between these two transparencies: $f(\mathcal{A} = 0) = 2/3$ and $f(\mathcal{A} = 1) \simeq 0.71$. The dependence of the current on the parameters γ, eV, ω_p and ξ is given by the prefactor in Eq. (36).

For $\kappa\hbar\omega_p/2 < eV <|\Delta|$,

$$\langle I\rangle = \frac{e}{h\hbar}\frac{16\gamma^4}{(1+\gamma^4)^2}\frac{\omega_p}{2}(\kappa - \xi^{-1})g\left(\mathcal{A}, \left(\frac{2eV}{\hbar\omega_p} - \xi^{-1}\right)(\kappa - \xi^{-1})^{-1}\right), \tag{38}$$

where

$$\begin{aligned} g(x,y) = \left(\frac{y}{x}\right)\Bigg(&\ln\left|1+\frac{x}{2}\left(1-\frac{1}{y}\right)\right| - \ln\left|1-\frac{x}{2}\left(1-\frac{1}{y}\right)\right| \\ &- \sqrt{\frac{y(2+x)}{x}}\Big\{\ln\Big|1-\sqrt{\frac{x}{(2+x)y}}\Big| - \ln\Big|1+\sqrt{\frac{x}{(2+x)y}}\Big|\Big\} - 2\sqrt{\frac{y(2-x)}{x}}\arctan\left(\sqrt{\frac{x}{y(2-x)}}\right)\Bigg), \end{aligned} \tag{39}$$

For $\gamma = 1$, the behavior of $\langle I\rangle$ is essentially the same as that of linear theory [4], for arbitrary ω_p. These results are plotted in Fig. 6 (with the choice $\omega_p = \Delta/4\hbar$).

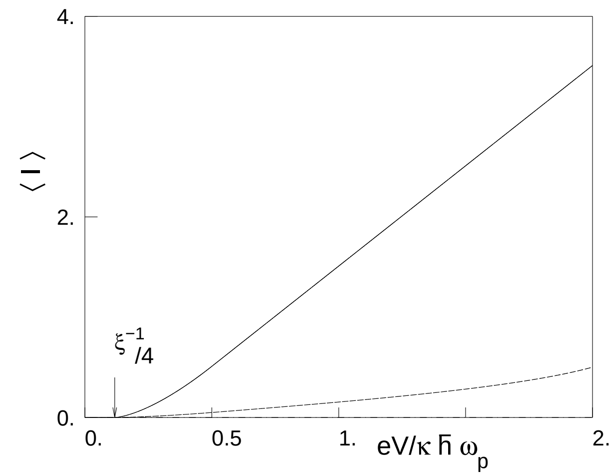

FIG. 6. Current $\langle I \rangle$ versus bias voltage in the disordered phase for $\hbar\omega_p/E_J = 2\pi$ [in units of $(4e\omega_p/\pi)\gamma^4/(1+\gamma^4)^2$]: perfectly good contact (solid line) and for $\gamma = 0.4$ (dashed line).

For eV larger than $\kappa\hbar\omega_p/2$, the curve $\langle I \rangle$ versus eV is a straight line. For less transparent barriers ($\gamma \neq 1$), two behaviors have to be distinguished. First, if the plasma frequency is smaller than the gap parameter, then the scaling variable $\mathcal{A}$ in Eq. (37b) is small and linear response is again retrieved. Second, if $\hbar\omega_p$ is comparable to $\mid \Delta \mid$, $\langle I \rangle$ increases typically as $(2eV - \kappa\hbar\omega_p\xi^{-1}/2)^{3/2}$ for $\xi^{-1}/2 < 2eV/\kappa\hbar\omega_p < 1$. Then, for eV larger than $\kappa\hbar\omega_p/2$, the response is non-linear. In particular, when the transparency is low, $\langle I \rangle$ increases faster than $(2eV/\kappa\hbar\omega_p - \xi^{-1}/2)^{3/2}$ for $eV > \kappa\hbar\omega_p/2$. Overall, no sensible deviations from linear response are expected in the disordered limit.

VI. CONCLUSION

A formulation of quantum transport in normal metal superconductor junctions is developed in the case where the order parameter undergoes phase fluctuations, both in the tunnel limit and in the transparent regime. The Keldysh Green's function was used, allowing to go beyond linear perturbation theory, provided that the subgap regime is specified (this latter assumption enabled us to neglect some contributions in the Dyson expansion). While we applied our results to the 1D Josephson junction array model of Ref. [4], the result is quite general as any model for the phase correlator can be employed to calculate the current and the finite frequency noise. An interesting issue is that the zero frequency noise satisfies the Schottky formula (as should be), with a current describing inelastic processes. A direct correspondence between the phase correlator and the second derivative of the noise spectral density with respect to frequency has been established. For a highly transparent interface, phase fluctuations are shown to affect only the differential conductance at low voltages: at high voltage the $I(V)$ characteristic is essentially that of a rigid superconductor, which can be described with the BTK model. On the other hand, for a less transparent barrier, deviations with respect to the BTK are shown to occur both at high and low voltages. This effect becomes more dramatic when the barrier is opaque.

The present system bears similarities with other situations where Andreev reflection is accompanied by the emission of a bosonic excitation. As mentioned in the introduction, Ref. [9] dealt with a ferromagnet whose magnons can flip the spin of one of two same spin electrons in order to contribute to the subgap conductance in a s-wave superconductor. Therefore, electrons with opposite spin contribute to the current as usual, but a new scattering channel – due to this one-magnon process – is opened, and enhances the Andreev current overall. The contrast with our calculation is that here, the current is always reduced by the excitation of phase modes. For a specified voltage bias, the more phase fluctuations, the more energetic are the modes which propagate in the Josephson junction array. On the side of the normal metal, this requires that the sum of the energies of the electron and hole has to be large. Yet the Fermi level restricts the impinging electron energy. This reduction of the available phase space for inelastic assisted Andreev reflection thus justifies a weaker current when the fluctuations are large. Also, note that the phase correlator which was used in this calculation typically appears in situations where a quantum mechanical degree of freedom is coupled to an electromagnetic environement [21]. Overall, if one includes multi-magnon processes to the situation of Ref. [9], both ferromagnetic/superconductor and normal-metal/fluctuating superconductor junctions constitute illustrations of quantum dissipative systems [22], where the quantum process to be probed – Andreev reflection – is coupled to a physical environment (a bath of magnons/"phase oscillators").

Potential applications to physical systems are now addressed. While typical BCS superconductors have essentially amplitude fluctuations (except close to the superconducting transition), the high-T_c cuprates have a much lower phase stiffness and hence, are expected to show strong fluctuations of the phase of the order parameter [23]. Here, an s-wave order parameter was assumed for simplicity, whereas these materials are mainly d-wave, but our calculation could in principle be generalized to this latter situation. The two main differences between s-wave and d-wave materials are the following: first, in a d-wave material, there are nodes in the order parameter in specific directions, which results in a finite density of states for quasiparticles near the superconductor chemical potential. Second, low energy Andreev bound states may exist at this energy. Depending on the orientation of momentum, an electron-like quasiparticle and a hole-like quasiparticle in the superconductor see an order parameter with the opposite sign [2]. This generates a zero-bias conductance peak (ZBCP) which is a hallmark of a d-wave order parameter.

At first sight, and for ideal interfaces, the connection of between our s-wave model and a d-wave compound could be made by first choosing the antinodal direction to be orthogonal to the plane of the interface [5], second by restricting electron transfer along the normal of this interface. Nevertheless, the confrontation of our theoretical results with transport experiments in high-T_c compounds may prove too difficult, as surface defects are likely to be present in such compounds. Such defects are responsible for a reduction of the order parameter in the vicinity of the junction. Moreover, the presence of Andreev bound states – also induced by the impurities at the interface – renders the present approach inadequate. Furthermore, surface roughness provokes a randomization of the tunneling directions so that the above scenario for antinodal tunneling loses its practicality.

Previous theoretical works addressing the role of phase fluctuations [24] on Andreev reflexion in the pseudogap phase [5,6] focussed on the mildly underdoped case: the hole density is chosen on the right hand side of the zero temperature insulator-superconductor quantum critical point (QCP). In this regime quantum fluctuations may renormalized the superfluid density but the physics turns out to be essentially that of the two-dimensional classical XY model (renormalized classical regime in the spirit of the $n = 2$, non-linear σ-model). Transverse superfluid velocity fluctuations due to the motion of vortices are expected to dominate. Because the time scales for the vortex motion, as measured by THz spectroscopy [25] are much larger than the Andreev time $\tau_A = \hbar/\Delta$, the correlation of the phase was treated as static (on the scale of τ_A). The phase correlator decays in space over a lengthscale ξ_{cl} which is the classical correlation length [16]. If tunneling occurs on an area which is much smaller than ξ_{cl}, then an Andreev signal is expected.

Our calculation (albeit in 1 dimension), focuses on quantum fluctuations. Fluctuations are included in a non-perturbative and systematic manner, regardless of the phase propagator which is used in the end. As a general trend, quantum fluctuations were found to have a tendency to reduce the Andreev reflexion signal. Note that other models of pseudogap also rely partly on quantum fluctuations [26], while others incorporate aspects of 1D physics (stripes) [27]. Extensions of our calculation beyond 1D – together with surface roughness – or with competing orders could be envisioned to achieve a more realistic description of STM experiments on high-T_c compounds.

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